UCRL-PROC-211062

LAWRENCE LIVERMORE NATIONAL LABORATORY

Magnetic Properties of Radiation Damage in Pu and Pu Alloys

S. McCall, M. J. Fluss, B. Chung, G. Chapline, M. McElfresh, D. Jackson, N. Baclet, L. Jolly, M. Dormeval

April 5, 2005

Actinides 2005
Manchester, United Kingdom
July 4, 2005 through July 8, 2005

Disclaimer

Magnetic Properties of Radiation Damage in Pu and Pu Alloys

S.McCall[1], M.J. Fluss[1], B. Chung[1], G. Chapline[1], M. McElfresh[1],
D. Jackson[1], N. Baclet[2], L. Jolly[2], and M. Dormeval[2]

[1]Lawrence Livermore National Laboratory, Livermore CA 94552 USA
[2]CEA-Centre de Valduc , 21120 Is sur Tille ,France

1 INTRODUCTION

Among the many exceptional properties of Pu[1] is its apparent lack of either local moments or cooperative magnetism. Lashley *et al.,*[2] have recently noted that little experimental evidence for the existence of local moments or collective magnetism has been found in over 50 years. Nevertheless the search for local moments in Pu and Pu-alloys continues[3], *why*? Plutonium's physical properties: resistance, magnetic susceptibility, and heat capacity[4], all support a system with an enhanced electron density of states. Pu sits on the edge of both magnetism and superconductivity and possesses one of the highest elemental Pauli susceptibilities, consistent with a highly correlated electron system. The low-density δ-Pu has eluded full first principles description and is both a challenge and an area of active investigation for theorists. The complex changes associated with the transition between the light and heavy actinides happen within the phase diagram of Pu, thus making Pu an intriguing and challenging solid-state system for continuing experimental and theoretical investigation.

Recently, Griveau *et al.,*[5] observed the variations in the resistance and superconducting properties of Am metal as a function of pressure to 27GPa and T>0.4K. They postulate that the interesting features in the superconducting critical temperature, T_c, *vs.* pressure, indicate a Mott-like, f-electron localization-delocalization transition as pressure drives Am towards a Pu and then a U-like structure. Hence, we posit that it would be reasonable to expect that dilating the Pu lattice will bring one to a similar transition. Experimental evidence supporting this point of view is given here.

2 PREVIOUS EXPERIMENTS

Recent experiments at Valduc and LLNL have pointed to the possibility of emerging magnetism with the accumulation of radiation damage. We briefly review these early experiments.

Dormeval describes an observation of the consequential effects of room temperature radiation damage accumulation in a series of Pu(Am) alloys[6]. The Am concentration, which increased the damage rate, ranged from 4.9% to 24%. The damage accumulation periods were such that each specimen had the same number of alpha decays. When the magnetic susceptibility of these iso-damaged specimens was measured (Figure 1 *left*) a feature was seen below 75K at fields below 2 Tesla that had not been present in the annealed specimen. What is interesting, however, is that this additional or excess magnetic susceptibility (EMS) could be significantly but not fully annealed at higher temperatures, indicating that radiation damage accumulation was indeed the source of the observed EMS. One expects that the accumulated damage consisted of helium-vacancy pairs, possibly small vacancy clusters, and the alpha decay products of Pu and Am, all of which may contribute to the EMS. In a related study, Fluss *et al.,*[7] observed a temperature dependence for the resistivity of vacancies and vacancy clusters, ρ, in Pu(3.3at%Ga) that obeyed a Kondo impurity form ρ=a-b*ln*(T) (Figure 1 *right*). This suggests that vacancies, while possessing no moment of their own, induce a Kondo-like moment in the surrounding material. An effect similar to this has been studied in the hole-doped superconductors[8] where zero spin impurities, including vacancies, in the Cu-O plane result in both Kondo behaviour and a reduction in T_c.

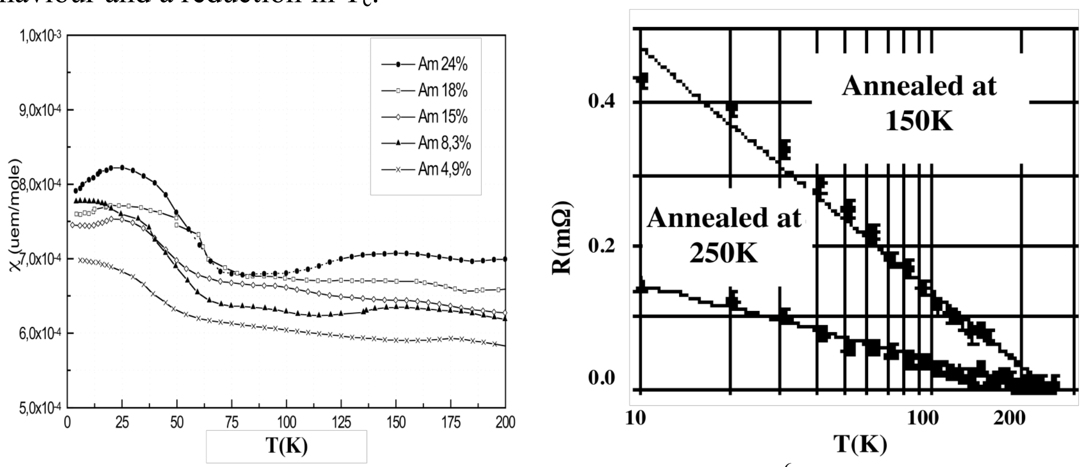

Figure 1 Left: *The appearance of EMS is seen in the data of Dormeval[6] for five iso-damaged specimens of Pu(Am).* Right*: The reported temperature dependence of the resistance of vacancy defects in Pu(3.3at%Ga) after 10K damage accumulation and either a 150K or 250K anneal[7].*

3 LOW TEMPERATURE DAMAGE ACCUMULATION

We report here preliminary data where we have measured the change in magnetic susceptibility as a function of time and temperature in α-Pu and δ-Pu(Ga). In terms of the atomic volume per atom α-Pu is close to Np while δ-Pu appears to be half way to Am. Hence, a comparison of the magnetic properties induced by accumulated radiation damage may be sensitive to this quantity. Isochronal magnetization annealing was used to determine the specimen temperature below which there was no defect mobility. Beginning with an annealed specimen lowered below the Stage I temperature, the magnetization was measured as a function of time and temperature while cycling from 2-30K, remaining safely below Stage I. In this way isotherms were determined for the two specimens. Some of this data is shown in Figure 2, where it is observed that at early time the EMS increases linearly, but after a few days substantial curvature evolves. The measured susceptibility χ(t,T) appears to fit a saturation model of the form in eq. 1, where χ_{Initial}(T) is the annealed susceptibility, χ_{EMS} is the maximum excess susceptibility, and τ(T) is a characteristic time.

$$\chi(t,T) = \chi_{Initial}(T) + \chi_{EMS}(T)(1 - e^{-t/\tau(T)}) \qquad (1)$$

What is noteworthy is that 1/τ(T) is proportional to an effective volume per alpha decay, which is the volume surrounding the U recoil damage cascade that has been modified; *i.e.* the volume in which the magnetic susceptibility has been increased by the local disorder. This volume for α and δ is 8 to 10 times greater than the circumscribed volume containing the damage cascade as estimated from molecular dynamics models. Delta-Pu reaches saturation sooner than α-Pu, meaning that the magnetically perturbed volume for δ-Pu is larger. This might be indicative of the concomitant increased localization from α– to δ–Pu.

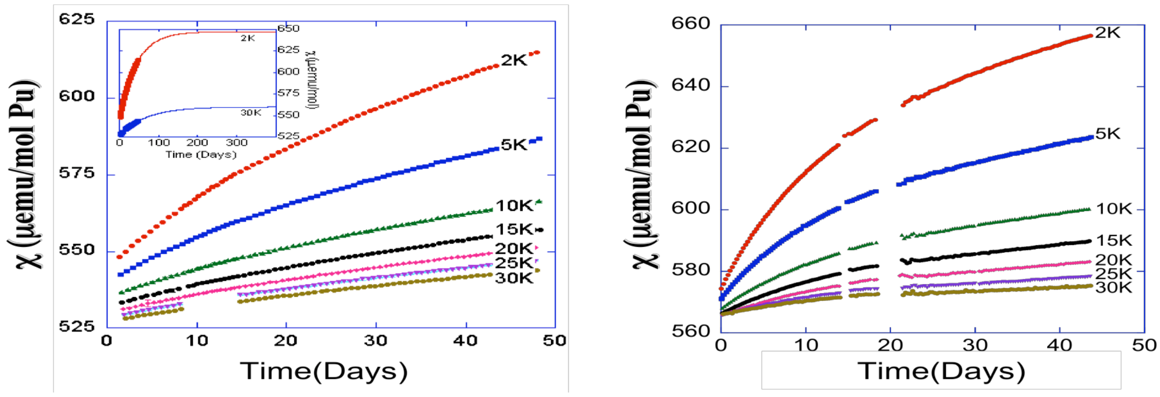

Figure 2 Left: *time evolution of magnetic susceptibility in α-Pu* Right: *and δ-Pu(Ga) each measured in a 3T applied magnetic field. The inset projects a fit of equation (1) to a year.*

4 CONCLUSION and FUTURE WORK

The exact structure-property relationships for the observed EMS described above remains as future work. However, we expect that vacancies, which create open regions in the lattice, should lead to increased f-electron localization via lattice dilation. It remains to be seen what contribution interstitials and interstitial loops make to the EMS. Dormeval[6] noted that in the case of Pu(Am), Am being oversized in the Pu lattice (positive deviation from Vegard's law) exhibits a clear anomaly in both susceptibility and resistivity at Am 25% (Figure 3). One might expect that low temperature damage accumulation studies close to this anomaly could lead to evidence for a delocalization to localization transition analogous to that seen in Am under pressure.

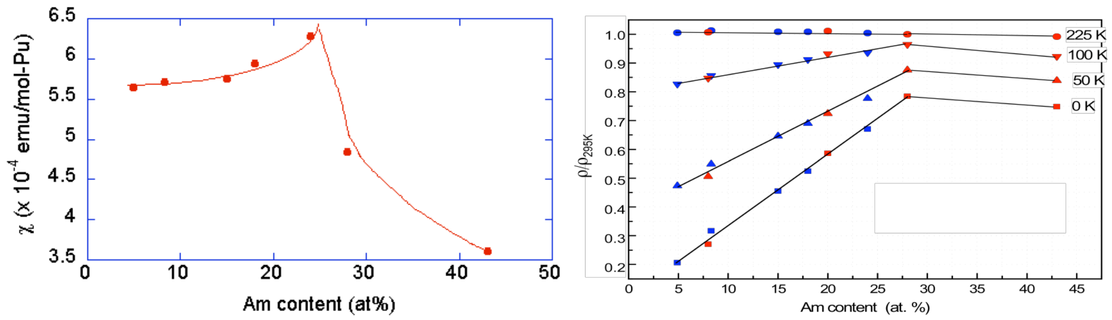

Figure 3 Left: *The Pauli susceptibility, χ_o, of Pu(Am) as a function of %Am, taken from(6).* Right: *Normalized resistive isotherms for Pu(Am) from(6) Both figures indicate an interesting discontinuity at ~25% Am suggesting increased localization. The lines are guides to the eye.*

References

1 Challenges in Plutonium Science, Los Alamos Science, Volume 26, I&II (2000), LA-UR-00-4100, edited by N. Cooper http://www.fas.org/sgp/othergov/doe/lanl/pubs/number26.htm
2 J.C. Lashley, *et al., submitted to*: Phys Rev B (LA-UR-04-3439) (2005).
3 S. Méot-Raymond and J.-M. Fournier, J. Alloys and Compds. **232,** (1996).
4 J. C. Lashley, *et al.,* Phys. Rev. Lett. **91**, 205901 (2003).
5 J.-C. Griveau, J. Rebizant, and G.H. Lander, and G. Kotliar Phys Rev Lett, *in press* (2005).
6 M. Dormeval, PhD Thesis, Universite de Bourgogne, Dijon, France, (2001). *and* N. Baclet, M. Dormeval, *et al*., J. of Nuc. Sci and Tech., Supp. 3, p148-151, (2002).
7 M.J. Fluss, *et al.*, J. Alloys and Compds. **368,** (2004).
8 *see for example and references therein:* F. Rullier-Albenque, H. Alloul, R. Tourbot, *Phys. Rev Lett.*, **91**(4), 047001, (2003).

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.